X-ray Scattering Studies Of The Hydration Kinetics In Tricalcium And Dicalcium Silicate Pastes

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Introduction: Cement is one of the most widely used construction materials, yet there is only a basic understanding of the hydration reaction that governs many of the final properties such as durability and strength. Tricalcium silicate (C_3S) is the most abundant phase in cement clinker, and thus represents the most simplified model for understanding the hydration reaction in cement. The reaction between C_3S and water is generally written: C_3S + (3+y-x)H --> C_xSH_y+ (3-x)CH, where (C = Ca^0, S = SiO_2, and H = H_2O) where x determines both the number of moles of calcium hydroxide (CH) formed and the C/S ratio of the C-S-H gel. By monitoring three regions of the spectra corresponding to the crystalline C_3S, crystalline CH and a region having peaks from both C_3S and CH we are able to monitor the progress of the reaction in real time. Three properties of the hydration reaction are being studied: temperature dependence, effect of particle size distribution, and the addition of varying amounts of C_2S to the reaction. Results from x-ray scattering are combined with data from inelastic and quasi-elastic neutron scattering to give a complete picture of they hydration reaction.

Methods and Materials: Initially pure C_3S will be used to calibrate the x-ray and neutron diffraction data. Then the effects of temperature will be studies at 10, 20, 30 and 40 °C. Additionally we have four chemically identical C_3S samples with different particle sizes centered around 1-2 μm, 10-12 μm, 25-30 μm, and 50-325 μm. Additionally, in an effort to understand the interaction of the different phases in real cement clinker, we will study samples with different ratios of C_3S and C_2S. A standard hydration reaction consists of mixing water and C_3S with a water/cement ratio of 0.4. The sample is then placed in a sealed container and placed in the beam in transmission mode. Data is collected over three regions with one data point every 0.01° and counting for six seconds per data point. This allows each region to be scanned in ten minutes, resulting in each regions being scanned once every 0.5 hours. Data was collected for 24-30 hours total at which point the reaction has slowed drastically.

Results: So far data has been collected only for one standard C_3S sample that has been well characterized using neutron diffraction techniques. **Figure 1** shows the resulting x-ray diffraction patterns at the start of the experiment and after 24 hours have elapsed. The difference between the spectra shows the decrease in the concentration of C3S and the formation of crystalline $Ca(OH)_2$. Data analysis generally consists of calculating the change in the concentration of the starting material C_3S and the formation of $Ca(OH)_2$. **Figure 2** shows the results from the two x-ray diffraction trials we performed on identical samples, compared with the results from neutron diffraction experiments take at two temperatures, 20°C and 30°C. Using x-ray diffraction we are able to measure the amount of C_3S which has reacted and the amount of $Ca(OH)_2$ which has formed (not show in graph) and compare this with the neutron diffraction experiments where we measure the fraction of water which has reacted.

Conclusions: Our initial results show that using x-ray diffraction to monitor the hydration kinetics in C_3S will provide additional information about the reaction primarily the rate at which C_3S disappears and $Ca(OH)_2$ is formed. This information is complementary to the neutron diffraction information allowing a more complete and detailed understanding of the hydration reaction. The results however also indicate that there was some difficulty in controlling the temperature at which the reaction occurs and possibly in reproducing the same sample thickness. We are currently redesigning our sample cell to have better temperature control and create a more reproducible sample thickness.

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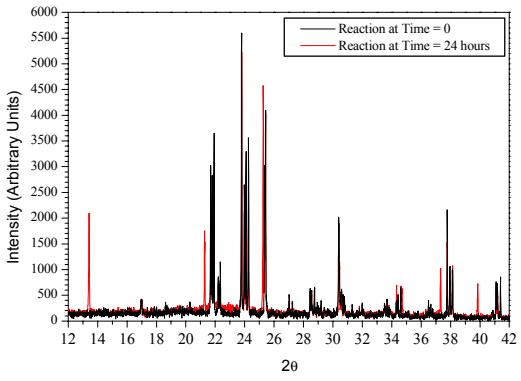

Figure 1. X-ray diffraction spectra at the start of the hydration reaction and after 24 hours.

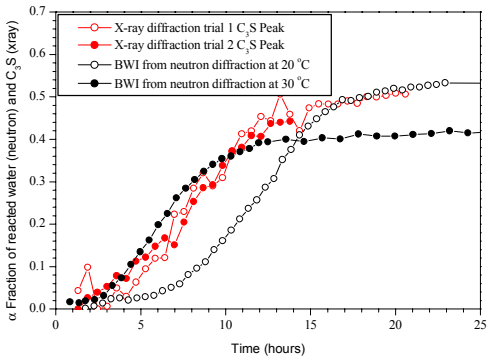

Figure 2a and b. Comparison of x-ray and neutron diffraction results. The x-ray data is for two trials on different samples to show the reproducibility of the method. The neutron data has been taken at two temperatures 20°C and 30°C